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Remote Sensing of Ozone Using an Infrared Differential Absorption System

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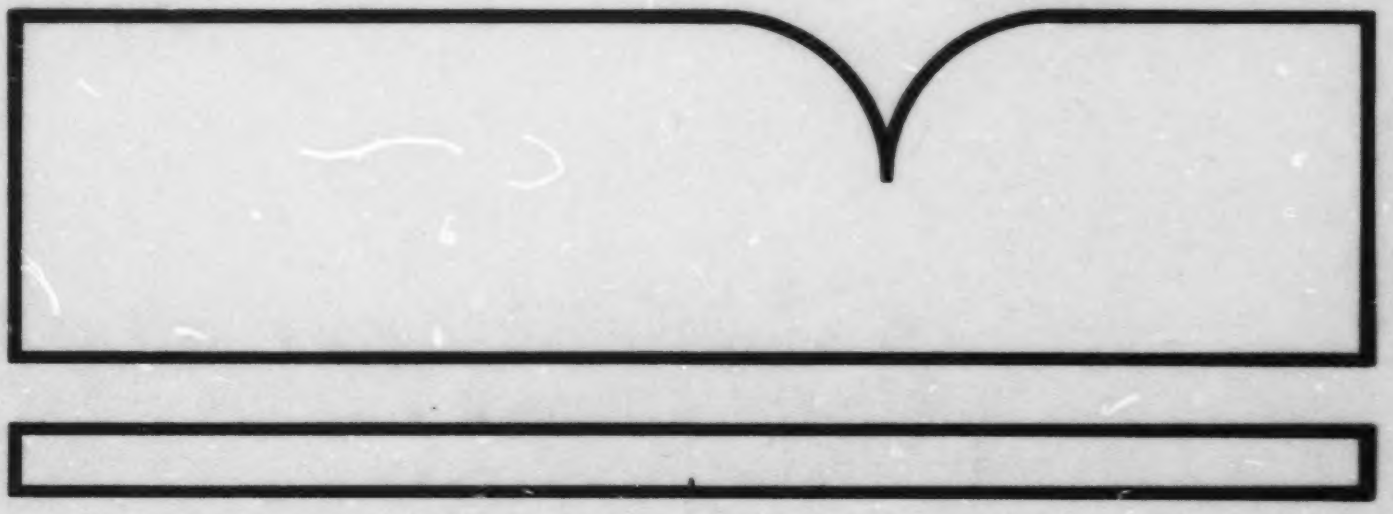

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16. ABSTRACT

A prototype airborne downlooking infrared differential absorption system using CO_2 TEA (transverse excited atmospheric) lasers is described. The system uses two wavelengths and topographic reflection to measure the integrated column concentration of ozone between the laser source/receiver and a noncooperative target. A comparison is made between ozone absorption coefficients measured with TEA lasers and values reported from other sources. Ground tests utilized two 30-cm long ozone-filled test cells, one in each laser path. A correlation was observed between measurements of TEA laser pulses backscattered from a building and ultraviolet determination of ozone concentration in the cells.

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REMOTE SENSING OF OZONE USING AN INFRARED DIFFERENTIAL ABSORPTION SYSTEM

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NOTICE

The authors were affiliated with the Environmental Monitoring Systems Laboratory when they worked on the infrared differential absorption system for remote sensing of ozone. As of the date of publication, J. L. Guagliardo is with Computer Genetics Corporation of Wakefield, Massachusetts; R. T. Thompson, Jr., is on assignment to the U.S. Environmental Protection Agency from Old Dominion University in Norfolk, Virginia; D. H. Bundy is with the Environmental Monitoring Systems Laboratory in Las Vegas, Nevada; and M. H. Wells is with the Nevada Power Company in Las Vegas, Nevada.

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INTRODUCTION

It was concluded by Kildal and Byer[1] that the resonance absorption technique is a far more sensitive method for remote measurement of atmospheric pollutants than either Raman or resonance backscatter methods. Byer and Garbuny[2] proposed the use of topographic targets as retroreflectors and stated that single-ended absorption methods with ranges of up to 10 km and sensitivities of less than 0.01 ppm were possible. This possibility has been further analyzed by Seals and Bair.[3] Menzies and Shumate[4] and Murray, et al.[5], have proposed methods in which differential absorption and topographic reflection are utilized for the measurement of air pollutants.

The method we describe ultimately is one in which the output from the CO_2 TEA lasers, tuned to two different wavelengths, is pulsed downward from an aircraft. The differential attenuation of the two pulses at different wavelengths reflected from a topographic target is then evaluated to infer the integrated concentration of ozone in a column between the aircraft and the local topographic feature.

We will use the differential absorption ozone device to study oxidant/precursor transport problems. Oxidant or oxidant precursor transport over distances on the order of 100 miles or more has been observed in studies such as those of Zeller et al.[6] and Spicer et al.[7] In order to develop appropriate control strategies, the actual magnitude of the impact of the transported oxidant and/or precursors must be quantified, and if possible, a model developed to relate emissions to their distant impact. Data from the earth-reflected differential absorption device, when coupled with windspeed, can be used to determine ozone flux from one area to another, a measurement which can only be approximated by other methods.

COINCIDENCE MEASUREMENTS

The prototype device was designed to utilize the high energy pulses available from TEA lasers. Most studies of the wavelength coincidence between CO_2 laser lines and ozone absorption lines employ lower pressure (2 kPa to 4 kPa) lasers,[8,9,10] whereas the TEA lasers operate at atmospheric pressure (≈100 kPa). Since the gain lines in the latter case are broader than those in the former, coincidence measurements were undertaken to investigate the suitability of the TEA lasers for use in a differential absorption system. Although the results presented here cannot be applied to all CO_2 TEA lasers, they are probably representative of what is to be expected.

The lasers employed in the laboratory and in ground field testing were grating tuned flowing gas CO_2 lasers, with outputs of 250 millijoules (at 10.5 μm) and a pulse duration of 250 ns, manufactured by GEN-TEC (model

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DDL-2SH). The ozone cells employed were made from 30-cm PVC tubing with IRTRAN II windows 25 mm in diameter. The laser pulses propagated through the cells were detected by GEN-TEC ED-100 power monitors. An ozone/oxygen mixture from a glass corona discharge cell was passed through the cells and the ozone concentration was monitored by ultraviolet absorption.

Results of the wavelength coincidence measurements are presented in Figure 1. The lasers were tuned over the P branch of the 9.4-μm CO_2 band. The measured attenuation coefficients for ozone absorption obtained with the TEA lasers are plotted for comparison with the calculated and measured values reported by Patty et al.[8] and Shewchun et al.[9]

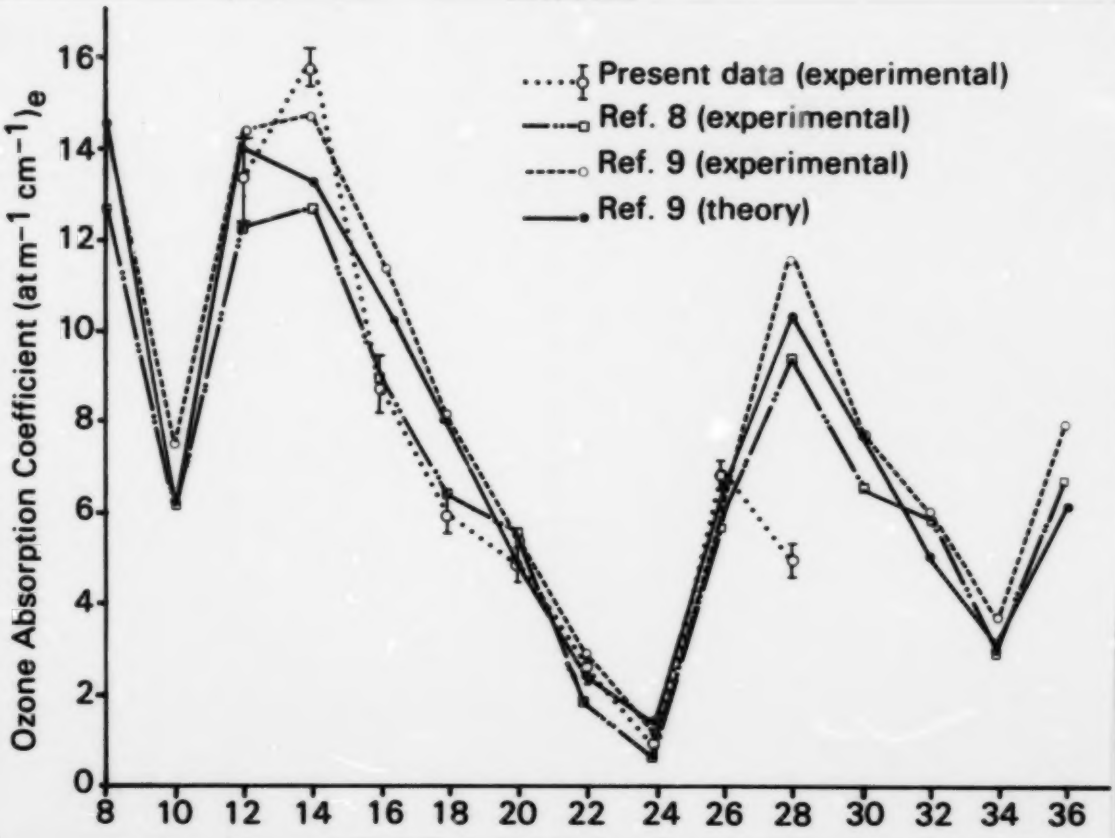

Figure 1. A comparison of experimental and theoretical values of ozone absorption coefficient at several CO_2 laser lines identified by J-value for the 9.4 μm band P-branch.

The measured attenuation coefficients were of the same order as those reported for lower pressure lasers. However, the variation of 20 percent to 50 percent at P(14) and P(28) by different observers could be attributed to variations in coincidence of a cavity mode controlled laser line position with the broadened ozone absorption lines. It is anticipated that detailed measurements would support the following argument.

There are several ozone absorption lines (≈7 within ±0.1 cm^{-1}) in the vicinity of the CO_2 P(14) and P(28) laser lines.[9,11] In general, TEA lasers will lase at several wavelengths under the gain profile as determined by the laser cavity parameters. When the pressure-broadened absorption coefficient of ozone due to all absorption lines in the vicinity of a given laser line is compared with the pressure-broadened gain profile of the laser, one expects the observed absorption coefficient to be strongly dependent upon the active modes of the laser within that gain profile. It is this type of limited wavelength coincidence which could lead to variations of the magnitude observed in Figure 1.

It is possible to stabilize laser cavity modes by introducing a saturable absorber[12], injection locking[13], or some other technique. An alternative to mode locking the laser is to incorporate an ozone absorption cell[14] in the system which would use a part of each transmitted laser pulse (<1 percent) obtained from a beam splitter to constantly monitor the effective ozone absorption coefficient.

In view of the apparently strong CO_2 J value dependence of the ozone absorption, the anticipated stability enhancement available through mode locking and the constant calibration updating available through the use of an ancillary absorption cell, it would appear that TEA lasers are suitable for use in an earth-reflected differential absorption system for monitoring ozone.

GROUND TESTING

THEORY

The backscattered radiant power from a Lambertian reflector is given by:[2]

$$P_r(R) = K\rho\frac{A}{\pi R^2}P_0 \exp[-2_0\int^R \xi_a(r)dr], \qquad (1)$$

where K is the optical efficiency of the receiving telescope (including overlap of laser field on telescope field), ρ is the reflectivity of the diffuse target, A is the telescope area, R is the distance to the target, P_0 is the transmitted power, and $\xi_a(r)$ is the volume attenuation coefficient, which is the sum of terms due to atmospheric scattering and absorption and to pollutant absorption.

If we consider measurements at two adjacent wavelengths and assume that atmospheric attenuation, target reflectivity, telescope optical efficiency, etc. are equal at both wavelengths, then the average concentration over distance R of pollutant C is:[15]

$$[C] = (\ln\frac{P_2 P_{01}}{P_1 P_{02}})/2R\Delta\alpha_c, \quad (2)$$

where P_n ($n = 1,2$) is the backscattered peak power at wavelength λ_n and P_{0n} is the transmitted peak power on wavelength λ_n, and $\Delta\alpha_c$ is the difference in the absorption coefficients of pollutant C measured at each of the two wavelengths.

A real-time signal processing system based on Equation (2) was designed and built[16] with values for backscattered peak power and transmitted peak power levels obtained from the system detectors. The R distance is obtained by timing the ground return, and the absorption coefficients are obtained through independent laboratory measurements. The processing system proved unsuitable for use in the EMI (electromagnetic interference) noise environment produced by the lasers, so detector response voltages were collected and processed subsequent to the experiment.

APPARATUS

A block diagram of the apparatus is shown in Figure 2. The lasers were identical to those used in the coincidence studies except that they were modified to utilize spark gaps and trigger generators purchased from a different manufacturer. The lasers and telescope assembly were isolated from the signal processing equipment by fiber optic LED's, photodiodes and cable. A pulse generator was used in a double pulse mode to trigger the waveform digitizer and to trigger the lasers 20 μs apart. The 20 μs is slow enough for double path round trip time for nonmultiplexed recording but fast compared to atmospheric scintillation and aircraft groundspeed. The zero-order reflection from the grating in each laser was detected by a fast (≈4 MHz) pyroelectric detector and used to measure the instantaneous power output of the laser for each pulse. The return pulse was collected by a 32 cm f/4 Newtonian telescope and focused onto a HgCdTe-LN_2 cooled photoconductive detector. The four pulses were digitized by a Biomation 8100 waveform digitizer and displayed on an oscillographic strip chart recorder.

A 30-cm cell was placed in front of each laser. The beams transmitted through the cells were allowed to propagate over a distance of about 300 meters to be diffusely reflected from the side of a building. Figure 3 shows the apparatus mounted on a test cart for this purpose.

The same corona discharge ozone generator as used in the coincidence measurements was used to alternately flow pure oxygen and oxygen/ozone mixtures through the cells. The ratio of ozone to oxygen was controlled by

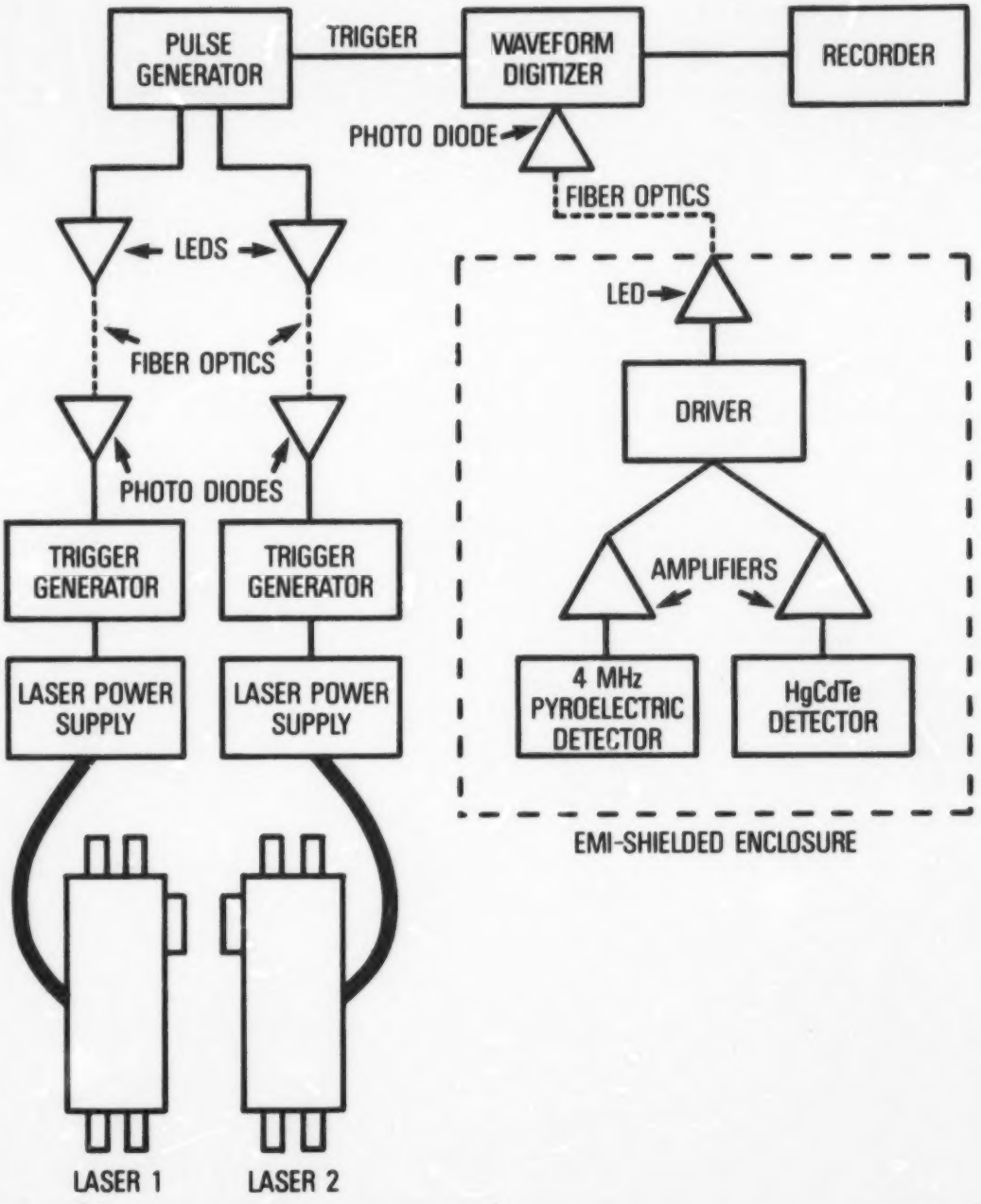

Figure 2. Schematic diagram of the system used for ground testing of differential absorption using a topographic reflector.

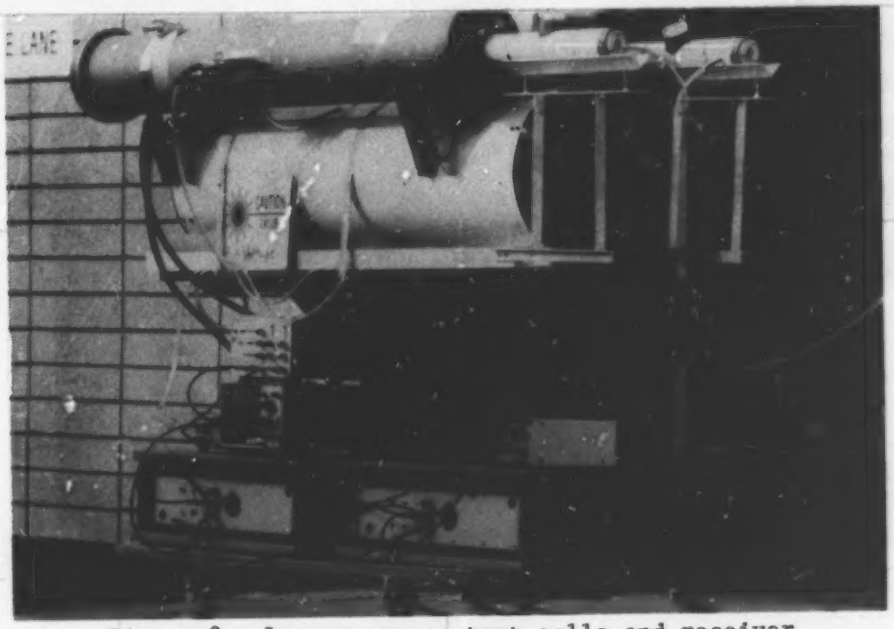

Figure 3. Lasers, ozone test cells and receiver telescope used for the ground tests are shown in this photograph.

varying the oxygen flow rate. The concentration of ozone in each cell was determined by measuring the change in ultraviolet absorption through a 10-cm quartz cell placed in series between the infrared absorption cells. The 254-nm line of a mercury pen-ray lamp isolated by a Shoeffel monochromator was used to measure the ozone concentration in the 10-cm quartz cell. Ambient ozone concentration was measured with a Dasibi ozone monitor.

RESULTS

A representative ozone measurement data set is shown in Figure 4. In particular, Figure 4 displays the noise, N_n, picked up from each laser discharge, n, followed in approximately 8 microseconds by a signal, P_{0n}, proportional in amplitude to the laser output energy and, in approximately 3 more microseconds, by a signal, P_n, proportional in amplitude to light backscattered from a topographic feature. The two lasers were tuned to wavelengths of λ_1 = 9.584 μm (P24) and λ_2 = 9.501 μm (P14).

A comparison of data obtained with the differential absorption system to those obtained by the ultraviolet absorption method is shown in Figure 5. Each data point in Figure 5 is computed from an average of 10 four-peak data sets similar to that shown in Figure 4. The cells were purged with oxygen before and after each variation in oxygen flow rate (ozone concentration is, within limits, inversely proportional to oxygen flow rate through the ozone generator).

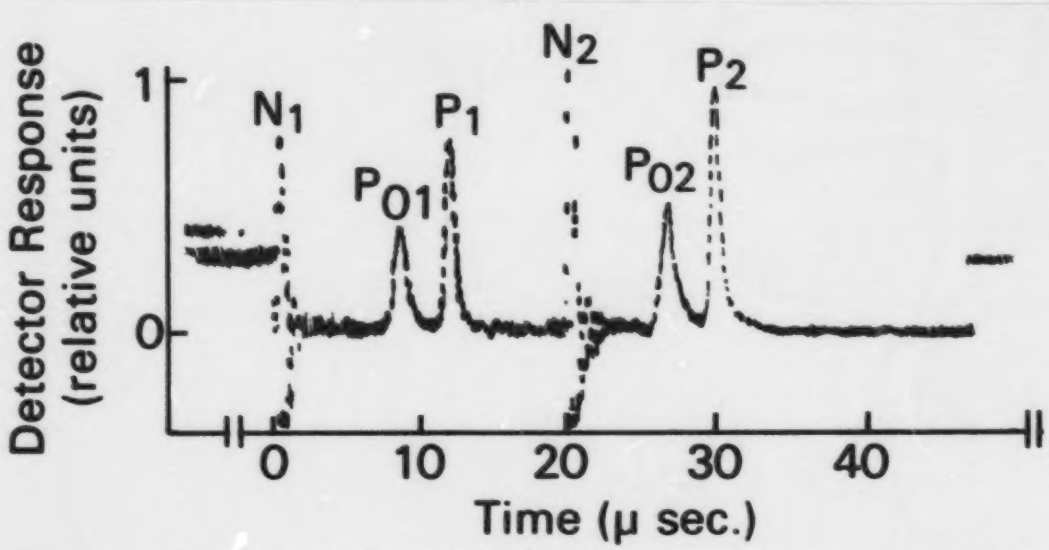

Figure 4. Description of a data set from a pair of laser firings where N_1, N_2 is the noise generated by the initial laser discharge, P_{01}, P_{02} is the power monitor response, and P_1, P_2 is the measured strength of the signal back-scattered from the target for laser 1 and laser 2 respectively.

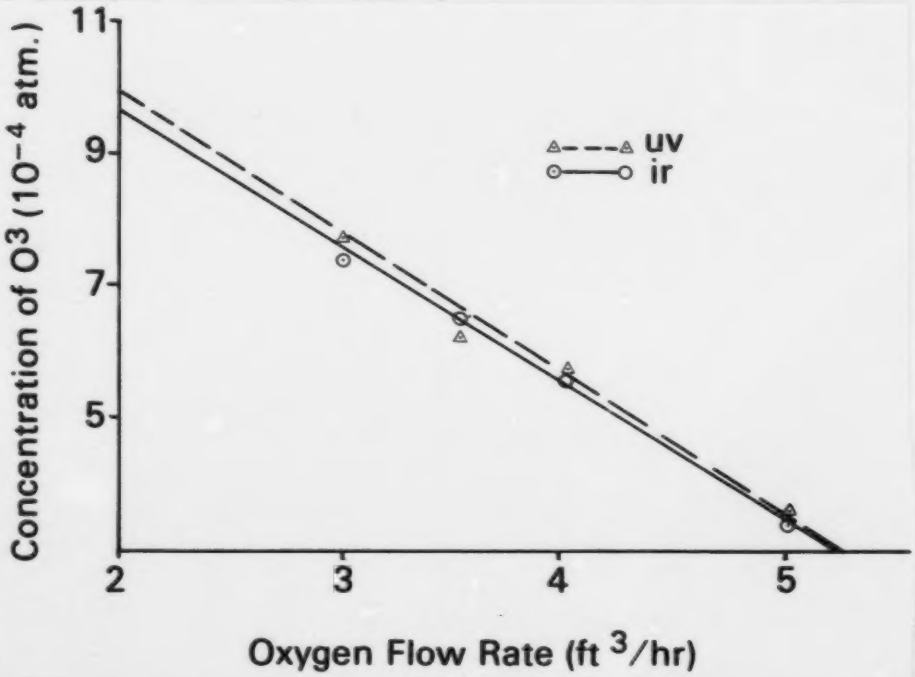

Figure 5. A comparison of ozone concentration in test cells as measured by differential IR absorption to that measured by UV absorption.

Dasibi readings indicated that ambient ozone levels averaged about 21 ppb during the course of the experiment. Ambient ozone accounted for about 1 percent additional infrared absorption over the pathlength between source/receiver and topographic target.

The variation between each set of four peaks, i.e., the results of each $\ln[(P_2P_{01}/P_1P_{02})]$, was only 5 percent without the cells in place. However, the variation was 20 percent at best with the cells in place, even when the cells were purged with nitrogen and the alignment maximized. This variation is thought to be due to a combination of interference effects and the fact that the telescope field of view was less than the divergence of the lasers (0.8 and 5 milliradians, respectively).

In addition, the lasers were known to operate in multiple high order transverse modes, as was indicated by several output burn patterns obtained on carbon paper. Modifications planned for the prototype should eliminate these problems by establishing mode control on the lasers, combining both beams in order to make the system coaxial, using a beam expander to control divergence and by using Brewster angled windows in the cell to eliminate interference effects.

CONCLUSIONS

Ground tests of the initial system design have disclosed certain limitations, which have been eliminated for the next modification. In spite of serious limitations imposed by a strong EMI field, an existence of many active laser modes, a lack of divergence control of the laser and a cumbersome test environment for conducting controlled tests, the system was able to demonstrate a correlation between infrared measurements of ozone and reliable ultraviolet measurements as indicated in Figure 5. The measurements reported in Figure 5 show the system capable of measuring ozone concentrations from 300 to 800 ppm in a 30-cm cell when operating 300 meters from a topographic target. This would correspond to average concentrations of 0.15 to 0.4 ppm over the 300 meters. Additional tests are planned for the redesigned system to determine the range dependence and concentration dependence of sensitivity for an earth-reflected differential absorption system.

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